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(54) A SOURCE OF IONIZING RADIATION

(71) We, ORDENA TRUDOVOGO KRASNOGO ZNAMENI INSTITUT FIZIKI AKADEMII NAUK GRUZINSKOI SSR, of ulitsa Guramishvili 6, Tbilisi, Union of Soviet Socialist Republics, a Corporation organized and existing under the laws of the Union of Soviet Socialist Republics, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to an ionizing radiation source suitable for the calibration of multichannel gamma-radiation analyzers.

Ionizing radiation sources are currently available which consist of a radioactive material deposited onto a non-radioactive material substrate such as, for example, polyethylene film. After radioactive material deposition onto the film, the radiation source thus obtained is sealed by coating the radioactive material with another polyethylene film and the resulting structure is placed in an appropriate holder.

These sources suffer from the disadvantages of being inadequately robust and of having the radioactive material non-uniformly distributed over the cross-sectional area of the source. The latter stems from the method of manufacture of the radiation source which involves the evaporation of aqueous or aqueous alcoholic solutions of radioactive materials.

The method is also disadvantageous in that it is not suitable for producing a large number of radiation sources having the same content of a radioactive material, so that in the case of sandwiched radiation sources each source must be subjected to a laborious calibration procedure to establish its activity.

Ionizing radiation sources are also available which comprise a radioactive material distributed in a mass of a non-radioactive material, for example aluminium, the sources being notable for their high mechanical strength and for their uniform radioactive material distribution throughout the mass.

However, aluminium-based radiation

sources are disadvantageous in that aluminium is a relatively strong absorber of gamma radiation in the 0 to 300 keV range.

Moreover, the method of manufacturing such aluminium-base sources which comprises alloying aluminium with a radioactive material makes it only possible to obtain sources with a limited number of radioactive metals such as, for example, cobalt-60, manganese-54 and zinc-65.

Among currently available ionizing radiation sources mention should also be made of polymer-base sources comprising a mixture of a radioactive material with an epoxide resin, the radioactive materials used being metal oxides.

The polymer-base sources suffer from the disadvantage of having the radioactive material non-uniformly distributed over the cross-sectional area of a source. This is particularly pronounced where the content of the radioactive material is less than 10^{-3} g per gram of the epoxide resin used and thus excludes the possibility of producing sources of an identical specific activity expressed in radioactive decay events per gram per second.

Each ionizing radiation source of this type requires laborious individual calibration to determine its activity.

Where this type of source is employed as a standard for the calibration of relevant instruments, the aforementioned disadvantages are the cause of additional errors.

The previously proposed ionizing radiation sources have an essential limitation as far as the employment of said sources for calibrating gamma-spectrometers furnished with high-resolution detectors (about 0.5 to 5.0 keV) is concerned, since the sources invariably contain only one radioactive isotope as the radioactive material, thereby rendering the measurements involved in the calibration procedure excessively laborious and protracted.

According to the present invention there is provided an ionizing radiation source comprising a moulded and hardened composition including a phenol-aldehyde resin and a radio-

active material ionically or molecularly dispersed in said resin.

A resol type of phenol-formaldehyde resin may be used as the resin component of the composition.

A novolak type phenol-formaldehyde resin mixed with hexamethylenetetramine may be used as the resin component of the composition.

Alternatively, a resol type resorcinol-formaldehyde resin may be used as the resin component of the composition.

A mixture of 70—90 parts by weight of resol type phenol-formaldehyde resin and 10—30 parts by weight of novolak type phenol-benzaldehyde resin may be used as the resin component of the composition.

Preferably, a mixture of spectrally pure gamma-emitting isotopes are employed as the radioactive material ionically or molecularly dispersed in the composition.

The invention will now be described in more detail, by way of example only, with reference to the accompanying drawings, in which:—

Figure 1 shows the gamma radiation spectrum of cobalt-60 in which the source activity expressed in pulses per minute is plotted on the ordinate and the numbers of gamma-spectrometer channels are plotted on the abscissa;

Figure 2 shows the gamma radiation spectrum of cesium-134, in which the source activity expressed in pulses per minute is plotted on the ordinate and the numbers of gamma-spectrometer channels are plotted on the abscissa;

Figure 3 shows the gamma spectrum of a source containing a plurality of isotopes, in which the source activity expressed in pulses per minute is plotted on the ordinate and the numbers of gamma-spectrometer channels are plotted on the abscissa.

The resins listed above are noted for their high mechanical strength, glass-like state, ability to react with ions of diverse elements, and low specific weight, said properties of said resins being retained in the ionizing radiation sources manufactured from the resins in question.

Hence, the ionizing radiation sources exhibit high mechanical strength and require no additional sealing on account of their glass-like state.

Chemical bond formation between radioactive elements and the hydroxyl groups of the resin used provides for the uniform distribution of the radioactive isotopes throughout the source mass, inasmuch as the resin-isotopes mixture is a solid molecular solution, the radioactive material particles being ionically or molecularly dispersed and forming no thermodynamic phase of their own.

In flat sources, isotope distribution over the cross-sectional area is uniform due to the

formation of true, (i.e. ionic or molecular) solutions of radioactive isotope ions in the resin.

The composition, from which the present source is made, consists essentially of light elements, such as carbon, hydrogen and oxygen, the specific gravity of said resin being low, viz. from 1.1 to 1.3 g/cm³, so that radiation self-absorption in the source will be insignificant. Thus, when the ionizing radiation source contains isotopes that emit gamma quanta having the energy of 200 keV minimum, the coefficient of self-absorption in a source about 1 mm thick amounts to less than 0.1—0.2% and can, therefore, be disregarded. Where ionizing radiation sources incorporate beta-emitting isotopes, the magnitude of self-absorption inherent in a given source is determined for each specific energy of beta particles by the conventional technique.

The following Examples described by way of illustration the procedures employed for the preparation of selected ionizing radiation sources.

EXAMPLE 1

The ionizing radiation source is a solid molecular solution of radioactive isotopes Mn-54, Fe-59 and P-32 in resol-type phenol-formaldehyde resin.

The procedure comprises irradiating iron sulphate in the neutron field of a nuclear reactor with a fast neutron integral flux of about 10¹⁸ neutrons per sq. cm., followed mixing 1 g of the irradiated iron sulphate with 10 mg. of radioactive manganese sulphate and 1 mg. of sodium dihydrogen phosphate containing a radioactive isotope of phosphorus. The resulting mixture is dissolved in 100 ml. of 96% ethanol, and the solution is introduced into a flask containing 200 ml of a 50% solution of resol-type phenol-formaldehyde resin in ethanol. Both solutions are mixed and the ethanol is thereafter distilled off in vacuo (ca. 50 mm of Hg) at a temperature of about 60°C on the water bath, followed by raising the temperature to 90°C and maintaining it for a period of 15 to 20 minutes to remove the water.

The thus-obtained syrup-like mass is placed in a vacuum thermostat and maintained under a vacuum of about 50 mm of Hg and at a temperature of about 80°C for 2 to 3 hours to yield a porous mass which is cooled to room temperature and ground to powder. The powder is tabletted under a pressure of from 250 to 1,000 kg/cm² and the resulting tablets 10 mm. in dia. and 1 mm. thick are heated at 80°C for 5 to 10 hours, the temperature being subsequently raised to 100—150°C and maintained for a period of from 2 to 5 hours to ultimately cure the composition.

The ionizing radiation sources obtained by the aforescribed procedure are noted for their high mechanical strength and find appli-

cation for the calibration of gamma spectrometers as to the energy of gamma quanta emitted by Mn-54 and Fe-59 and also of beta particles emitted by P-32.

5 EXAMPLE 2

The ionizing radiation source comprises a solid molecular solution of Co-60 in resol-type phenol-formaldehyde resin.

Shown in Figure 1 is the gamma spectrum of such a source taken on a multichannel analyzer having a semiconductor-type Ge(Li) detector (volume, 25 cm³). As can be seen from the spectrum, the source contains a pure isotope since the spectrum is free from gamma peaks due to impurities. The source can be used conveniently for gamma spectrometer calibration with respect to the energies of about 1,170 and 1,333 keV.

The procedure of preparing said gamma emitting source comprises dissolving 0.5 g of cobalt nitrate that contains radioactive isotope Co-60 and has an overall activity of 100 microcurie (mcC) in 50 ml. of 96% ethyl alcohol. Next the cobalt nitrate solution is introduced into a flask containing 200 ml. of a 50% solution of resol-type phenol-formaldehyde resin in ethyl alcohol, and the resulting mixture is processed as described in Example 1.

The ionizing radiation sources thus prepared are tablets 10 mm. in dia. and 1 mm thick, the activity of each source being about 0.1 mcC.

35 EXAMPLE 3

The ionizing radiation source, which consists of resorcinol-formaldehyde resin with radioactive isotope Cs-134 uniformly distributed therein, is prepared in accordance with the following procedure.

One gram of Cesium nitrate (overall activity, 100—500 mcC) is dissolved in 100—150 ml. of 96% ethyl alcohol, and the solution is introduced into a flask containing 200 ml. of a 50% solution of resol-type resorcinol-formaldehyde resin in ethyl alcohol, and the mixture is stirred.

Next the ethyl alcohol is stripped from the mixture by placing the flask on the water bath and heating the contents under vacuum (10—50 mm of Hg) at a temperature of about 60°C, followed by raising the temperature to 90°C and distilling off the water for a period of 15 to 20 min. The resulting syrup-like mass is next poured into dishes made of polytetrafluoroethylene and maintained in a thermostat at a temperature of from 80° to 100°C for a period of 24—48 hours to cause composition hardening.

When use is made of dishes ca. 10 mm in dia and 5 mm high, 1 kg of the resin is adequate for the manufacture of about 1,000 to 1,500 ionizing radiation sources, provided production losses are accounted for.

Figure 2 presents the gamma spectrum of the Cs-134 source plotted by means of a multichannel analyzer having a 25 cm³ Ge (Li) detector. As can be seen from the graph, the source contains one radioactive isotope only.

60 EXAMPLE 4

The ionizing radiation source comprises a solid molecular solution of Cs-134, Mn-54, Sc-46, Zn-65 and Co-60 in resol-type phenol-formaldehyde resin modified with 20% of novolak-type phenol-benzaldehyde resin.

The energy of photopeaks and the absolute activity of each isotope in the source are listed in Table 1.

TABLE 1
Energy of principal photopeaks, keV

Isotope	Activity, microcurie	E _{γ1}	E _{γ2}
Cs-134	0.15	604	796
Mn-54	0.25	835	—
Sc-46	0.30	890	1,119
Zn-65	0.30	1,110	—
Co-60	0.40	1,172	1,333

The following procedure is employed for the manufacture of this type of source.

The isotopes listed above are taken in the form of nitrates containing respective non-radioactive isotopes, and the mixture is prepared from 0.4 g of each salt, the activity of each salt being selected to be as follows: Co-60, 60-80 mcC; Zn-65, 60 mcC; Sc-46, 60 mcC; Mn-54, 50 mcC, and Cs-134, 30 mcC. The mixture is dissolved in 100 ml. of 96% ethyl alcohol.

Next 20 g of phenol-benzaldehyde novolak

and 80 g of resol-type phenol-formaldehyde resin are placed in a flask, 100 ml. of 96% ethyl alcohol is added thereto, the mixture is stirred until the resins undergo complete dissolution, and then the previously prepared alcoholic solution of the mixture of radioactive metal nitrates is introduced into the flask.

Stripping the alcohol and water from the contents of the flask is conducted as described in Examples 1 or 3.

The resulting syrup-like mass is poured, in

0.5-gram batches, into polytetrafluoroethylene dishes 15 mm in dia. which are placed in a thermostat and maintained at a temperature of 100°C for a period of from 48 to 72 hours.

5 The procedure described hereinabove yields ionizing radiation sources noted for their identical content of radioactive isotopes which corresponds to the composition listed in Table 1.

10 The gamma spectrum of this type of source is shown in Figure 3. As can be seen, the resolving power of the Ge(Li) detector used is adequate for discriminating all the photopeaks, so that measurements involving the use of one
15 such source only provide data which are sufficient for the energy calibration of multichannel analyzers.

EXAMPLE 5

20 The ionizing radiation source comprises a composition based on phenol-formaldehyde novolak cured by means of hexamethylenetetramine and containing radioactive isotope Mn-54 distributed therein.

25 To prepare this type of source, use is made of the following procedure. To 5 ml. of an aqueous solution of manganese sulphate containing only the Mn-54 isotope and no inactive carrier and having the total activity of 100 mcC is added 100 ml. of 96% ethyl alcohol, and the mixture is stirred, followed
30 by introducing said solution into 200 ml. of a 50% solution of phenol-formaldehyde novolak in ethyl alcohol, and thereafter adding to the mixture 100 ml. of the solution obtained by dissolving 10 g hexamethylenetetramine in ethyl alcohol. Next the mixture is thoroughly
35 mixed and thereafter processed in accordance with the procedure disclosed in Example 1.

40 The resulting gamma radiation sources have an activity of about 0.1 mcC. In all sources of a given lot, the specific activity of the composition, from which the sources are made, is constant to within 0.5%.

EXAMPLE 6

45 The ionizing radiation source comprises a hardened composition obtained from 60% of resol-type phenol-formaldehyde resin and 40% of novolak-type phenol-formaldehyde resin and having Co-58 distributed therein.

50 This type of source is prepared by the following procedure. To 10 ml. of an aqueous solution of cobalt-58 acetate having a total activity of 500 mcC there is added 0.5 ml. of an aqueous solution of non-radioactive cobalt acetate (concentration, 10^{-6} g./ml.) and also
55 100 ml. of 96% ethyl alcohol, followed by stirring the resulting solution and transferring it into a flask that contains 200 ml. of a solution of 60 g of resol-type phenol-formaldehyde resin and 40 g of novolak-type phenol-formaldehyde resin in ethyl alcohol.
60 The mixture is thoroughly stirred and thereafter processed by following the procedure of Example 4.

65 The activity of the resulting gamma radiation sources equals 0.5 microcurie, and in all sources of a given lot, the specific activity of the composition from which the sources are made is constant to within 0.5%.

70 In all instances where the sought-for ionizing radiation sources are produced by dispersing appropriate radioactive isotope salts in phenol-aldehyde resins, there occurs the formation of true solid solutions of said isotopes throughout the entire resin bulk, thereby making it possible to readily obtain a large number of sources having an absolutely identical specific content of a given radioactive isotope.

80 About 5,000 to 7,000 ionizing radiation sources in the form of tablets ca. 10 mm in dia. and ca. 1 mm thick can be manufactured from 1 kg of the phenolic resin, the radioactive standardization of the sources being substantially simplified by the fact that it is sufficient to determine the specific activity
85 of from 3 to 10 sources, while the activity of all other sources can be obtained with a high degree of accuracy by weighing precisely each source and making simple calculations to estimate the activity.

90 The ionizing radiation sources which contain several radioactive isotopes are eminently suited for calibrating multichannel gamma spectrometers furnished with semiconductor detectors of a high resolving power of about
95 0.5 to 5 keV.

100 The availability of ionizing radiation sources that contain a plurality of gamma-emitting isotopes provides the possibility of rendering the procedure of calibrating multichannel gamma analyzers less laborious and of enhancing the accuracy of measurements.

105 In the described ionizing source, the radioactive material used consists of any desired combination of gamma-emitting isotopes and the distribution of said isotopes over the cross-sectional area and through-out the mass of the source is uniform.

WHAT WE CLAIM IS:—

1. An ionizing radiation source comprising
110 a moulded and hardened composition including a phenol-aldehyde resin and a radioactive material ionically or molecularly dispersed in said resin.

2. An ionizing radiation source as claimed
115 in claim 1 wherein in said composition the phenol-aldehyde resin is resol-type phenol-formaldehyde resin.

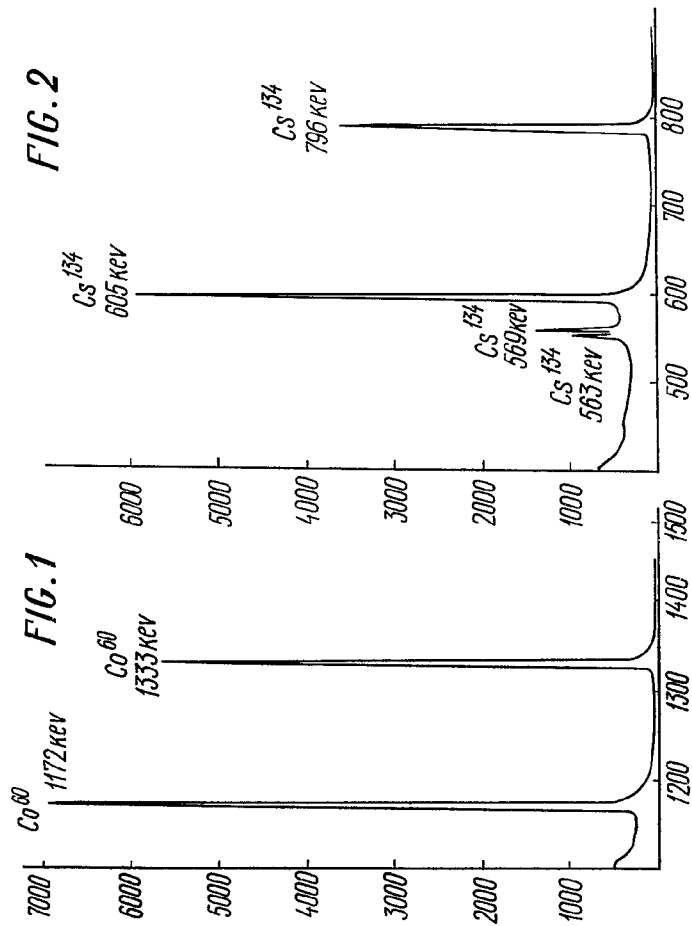
3. An ionizing radiation source as claimed
120 in claim 1 wherein in said composition the phenol-aldehyde resin is novolak-type phenol-formaldehyde resin admixed with hexamethylenetetramine.

4. An ionizing radiation source as claimed
125 in claim 1 wherein in said composition the phenol-aldehyde resin is resol-type resorcinol-formaldehyde resin.

5. An ionizing radiation source as claimed in claim 1 wherein in said composition the phenol-aldehyde resin is a mixture of 70 to 90 parts by weight of resol-type phenol-formaldehyde resin and 10—30 parts by weight of novolak-type phenol-benzaldehyde resin.
- 5 6. An ionizing radiation source as claimed in any one of claims 1 to 5 wherein in said composition the radioactive material dispersed
- 10 ionically or molecularly therein is a mixture of spectrally pure gamma-emitting isotopes.
7. An ionizing radiation source as claimed in any one of the preceding claims and substantially as herein described.

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COMPLETE SPECIFICATION

2 SHEETS

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Sheet 2

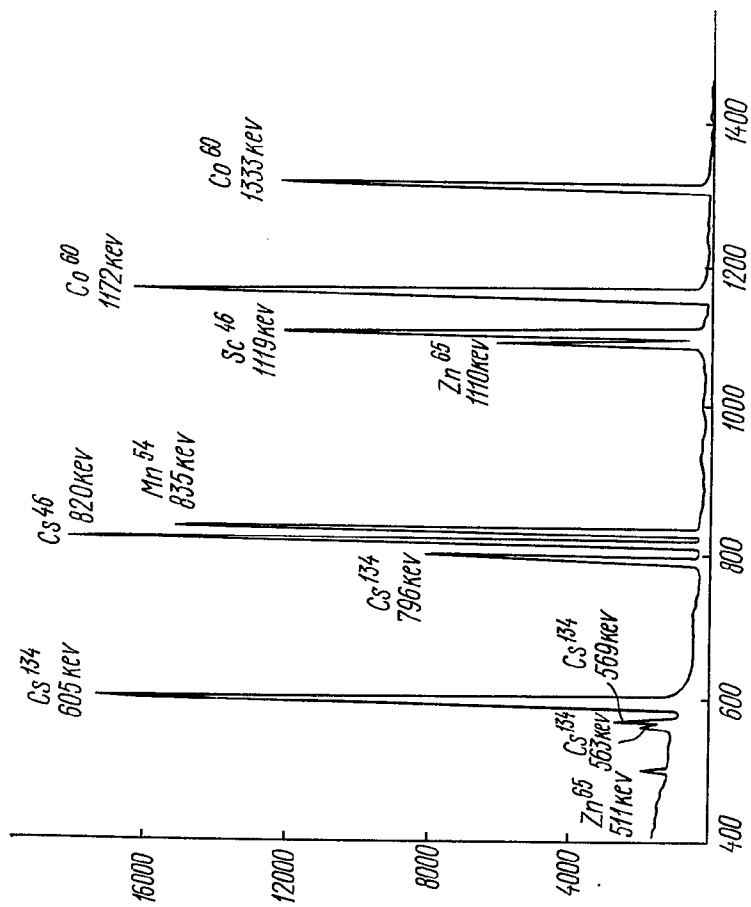


FIG. 3